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# SEPARATION OF A BINARY MIXTURE WITH A LARGE MASS DIFFERENCE BETWEEN THE COMPONENTS IN A GAS CENTRIFUGE

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*The separation of a binary gaseous mixture of uranium hexafluoride $^{238}UF_6$ with different light components in a high-speed centrifuge, intended for separating heavy isotopes, is examined. The mass of the light impurities is varied in the range $M_1$ = 0.02–0.349 kg/mole. It is shown that as the impurity mass decreases the structure of the flow fields in the centrifuge rotor changes considerably. If in the case of a mixture of heavy isotopes convective transport has a determining effect on the concentration, for a mixture with a large difference of the mass of the components radial barodiffusion in a centrifugal field becomes primary, which results in the appearance of a maximum of the separation factor at $M_1$ = 0.127 kg/mole.*

Centrifugal isotope separation is now used in the production of nuclear fuel from natural raw material, secondary processing of depleted uranium, spent fuel and weapons uranium, and stable isotope production. Depending on its origin, a gas mixture can contain not only relatively heavy target components but also a small quantity of elements with average and low mass, which above the admissible concentration disrupt the operation of equipment. For this reason, the calculation of the distribution of components inside the rotor of a centrifuge is a topical problem.

In general, the separation factor of a binary mixture with arbitrary mass of the components in a single gas centrifuge can be calculated using the equations of motion of a continuous medium and diffusion. In most analytic and numerical methods for calculating separation, the isotopic approximation or other limitations are used. A modified analytical method of averaging over the radius is developed in [1]; it takes account of the special nonuniformity of the transit flows. Numerical modeling of the separation of certain isotopic mixtures is done in [2], but these data are insufficient for analyzing the mass dependence of the separation factor. The separation of the mixture $UF_6$–$N_2$ in a low-speed rotor with the simplified circulation flow studied in [3] is inapplicable for modern high-speed centrifuges.

The following phenomenological dependence is proposed on the basis of a generalization of the computational data [4]:

$$\gamma_{ij} = \gamma_0^{M_j - M_i}, \tag{1}$$

where $\gamma_{ij}$ is the total separation factor of the centrifuge; $\gamma_0$ is the separation factor for unit mass difference of the components; and $M_i$ and $M_j$ are the molar mass of the $i$th and $j$th components. The separation factor for unit mass difference of the components is an adjustable parameter and reflects the effect of many physical factors: geometry of the centrifuge, rotational velocity and structure of the gas flow in the rotor and others. Relation (1) is often used in theoretical analysis of centrifugal separation of arbitrary multicomponent mixtures. The separation factor for unit mass difference of the components, as a rule, is assumed to be constant.

The objective of the present work is to study the separation of a binary mixture with a large mass difference between the components in a gas centrifuge, initially optimized for uranium isotope separation.

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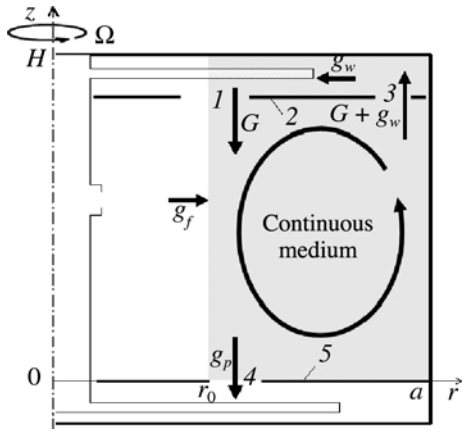


Fig. 1. Schematic diagram of the rotor of a gas centrifuge with the main structural elements: *1*) recirculation opening; *2*, *5*) waste and product diaphragms, respectively; *3*, *4*) peripheral and product openings, respectively.

We shall study stationary axisymmetric convective diffusion of a binary gas mixture $^{238}U_6$–imp (imp – light component, impurity) in the rotor of an Iguaçu centrifuge [5] with radius $a$ and height $H$. The mass of the impurity varies in the range $M_1 = 0.02$–$0.349$ kg/mole, the uranium hexafluoride mass is fixed $M_2 = 0.352$ kg/mole. The computational region includes the densest peripheral zones of the product and waste chambers (Fig. 1). In these zones, $Kn_{UF_6} = \lambda/l \ll 1$ is the Knudsen number, characterizing the rarefaction of the gas; $\lambda(r)$ is the average free path length at distance $r$ from the rotor axis; and $l(r) \sim 1/\rho(d\rho/dr)$ is a characteristic radial scale of variation of the density. The recirculation openings *1* of the waste diaphragm *2* and the openings *4* of the product diaphragm *5* are located in the transitional zone ($Kn_{UF_6} \sim 1$) near the inner boundary $r = r_0$ of the region under study. It is assumed that the pressure of the light component in the rarefaction zone of the gas $0 < r < r_0$ is low enough that its effect on the continuous medium $r_0 < r < a$ can be neglected.

The circulation flow in the rotor is optimized beforehand with respect to the separation of heavy isotopes $^{238}UF_6$–$^{235}UF_6$ by varying the temperature difference $\Delta T$ on the side wall of the rotor by adjusting the recirculation flow $G$ and the action of the waste flow separator. The latter is modeled by a volume mass sink and azimuthal angular momentum in the equations of motion of a continuous medium [6]. These parameters remain constant in the course of the calculations. The feed flow enters the dense region through its inner boundary $r = r_0$ in the form of a uniformly distributed flow in the interval $0.5 < z/H < 0.65$.

The exterior flows satisfy the balance equations for matter:

$$g_f = g_p + g_w,$$

where $g_f = g_{1f} + g_{2f}$, $g_p = g_{1p} + g_{2p}$, and $g_w = g_{1w} + g_{2w}$ are the mass feed, product and waste flows; $g_{1f}$, $g_{1p}$, and $g_{2w}$ are the mass flows of the lighter component in the feed, product and waste flows, respectively; $g_{2f}$, $g_{2p}$, and $g_{2w}$ are the mass flows of the heavy component in the feed, product and waste flows, respectively.

The mass concentration of the impurity in the feed flow $c_f = g_{1f}/g_f$, product flow $c_p = g_{1p}/g_p$, and waste flow $c_w = g_{1w}/g_w$ are related by the balance relation

$$c_f = \theta c_p + (1 - \theta)c_w.$$

The mass fraction (concentration) of the impurity in the feed flow is set so that the molar fraction (concentration) is the same (0.15%) for any impurity mass.

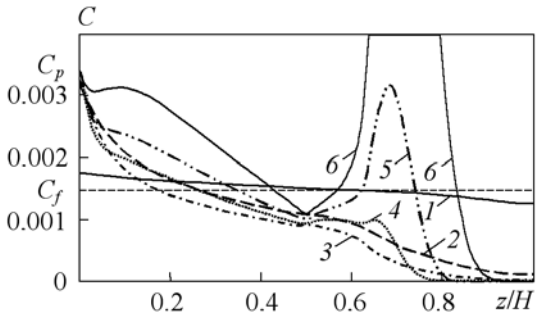


Fig. 2. Molar concentration of the impurity versus the axial coordinate at the boundary of the continuous medium ($r = r_0$, $r/a \sim 0.81$) for the mass difference of the components in the mixture $\Delta M = 0.003$ (*1*), 0.032 (*2*), 0.052 (*3*), 0.202 (*4*), 0.252 (*5*), and 0.332 (*6*).

The partition coefficient of the flow of the mixture is fixed, $\theta = g_p/g_f \sim 0.5$. The partition coefficient of the light flow

$$\theta_1 = g_{1p}/g_{1f} = \theta(c_p/c_f) \tag{2}$$

and the heavy component $\theta_2 = g_{2p}/g_{2f} = \theta(1 - c_p)/(1 - c_f)$ depends on the concentration and can vary as a function of the mass difference. For low impurity concentration $c \ll 1$, the partial partition coefficient of the flow of the heavy component is practically identical to the partition coefficient of the mixture flow $\theta_2 \sim \theta$.

The partition coefficient of the binary mixture is determined from the relation

$$\gamma = \frac{c_p}{1 - c_p} \Big/ \frac{c_w}{1 - c_w}.$$

**Numerical Computational Method.** The hydrodynamic parameters of the gas are determined from the equations of motion: Navier–Stokes, heat conduction and continuity, written in a cylindrical coordinate system in the approximation of axial symmetry. Since the impurity concentration is low, the viscosity and thermal conductivity for the mixture are almost identical to those of the heavy component. The concentration is determined by the equation of convective diffusion where the dependence of binary diffusion on the mass ratio of the light and heavy components is taken into account [7]:

$$D = D_0 \sqrt{\frac{M_2 + M_1}{2M_1}},$$

where $D_0$ is the self-diffusion coefficient for the heavy component.

The system of six coupled nonlinear partial differential equations with prescribed boundary conditions can be solved numerically by Newton's iterative method using a finite-difference approximation [8].

**Discussion.** It is evident in Fig. 2 that as the mass difference between the components is varied the concentration of the impurity in the lower enrichment part of the rotor varies much less than in the upper depletion part. In addition, in the upper part as the mass difference in the increases in the range $0.003 < \Delta M < 0.052$ kg/mole (curves *1*, *2*, *3*), the radial concentration differential is much smaller than the change of the concentration over the height of the rotor. The subsequent increase of the mass difference ($0.202 < \Delta M < 0.332$ kg/mole) is due to a sharp increase of the concentration (curves *4*, *5*, *6*), impurity redistribution occurs in the rotor, and for $M_1 < 0.12$ kg/mole the radial and axial concentration gradients become comparable to one another.

It is shown in Fig. 3 that for $\Delta M > 0.072$ kg/mole the partition coefficient of the impurity flow goes to 1. In accordance with relation (2), the largest partition coefficient of the flow is reached for $c_p \sim c_f/\theta$. In this case, almost all the light

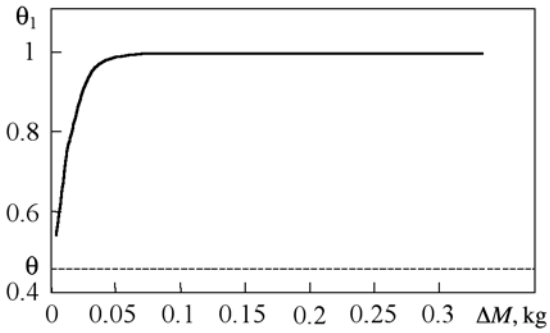


Fig. 3. The partition coefficient of the impurity flow versus the mass difference $\Delta M$ of the components of the mixture.

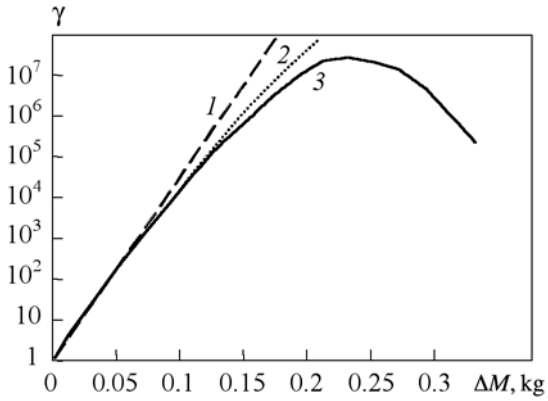


Fig. 4. The partition coefficient versus the mass difference of the mixture components $\Delta M$: *1*) expression (1) at $\gamma_0 = 1.11$; *2*) modified method of radial averaging [1]; *3*) results of numerical calculation.

component entering with the feed goes into the product. Since the partition coefficient of the mixture flow is kept constant ($\theta \sim 0.5$), the maximum possible impurity concentration in the product flow is determined from the expression $c_{p\,\max} \sim 2c_f$.

Figure 4 shows that for small mass differences between the components ($0.003 < \Delta M < 0.072$ kg/mole) the analytical (curve *2*) and numerical (curve *3*) methods give approximately the same results: the dependence of the partition coefficient on the mass difference is close to an exponential function (1) (curve *1*). As the mass difference increases, curves *2* and *3* deviate from an exponential. For large mass difference, function *2* remains monotonic, and a maximum appears in curve *3* near $\Delta M \sim 0.225$ kg/mole.

The appearance of a maximum of the partition coefficient can be explained as follows. As the mass difference increases, the partition coefficient of the flow increases initially, going to 1 (see Fig. 3). For $\theta_1 \sim 1$ the impurity concentration in the product flow is close to the limit $c_{p\,\max} \sim 2c_f$ and remains unchanged with subsequent increases of the mass difference. After this, for an even larger increase of the mass difference the partition coefficient changes only as a result of the variation of the impurity concentration in the waste flow. In the interval $0.072 < \Delta M < 0.225$ kg/mole radial barodiffision transfer of the light component is small relative to convective transfer in the axial direction, which results in a decrease of the waste concentration and increase of the partition coefficient. For $\Delta M > 0.225$ kg/mole, the radial transfer predominates over axial convective transfer of the light component, the impurity concentration in the depletion part of the rotor and the waste flow increases and the partition coefficient decreases.

In summary, it was shown by a direct numerical calculation that for components with close mass the dependence of the partition coefficient on the mass difference is close to an exponential function. For mass difference $\Delta M > 0.05$ kg/mole, almost all the impurity entering with the feed flow goes into the product flow. An extremum of the partition coefficient was found for the case of a significant mass difference between the components; this extremum is due to the fact that the radial transfer of the components predominates over axial transfer. For a centrifuge [5], the maximum partition coefficient $10^7$ is reached with partition of a uranium hexafluoride mixture with mass $M_2 = 0.352$ kg/mole with a component with mass $M_1 = 0.127$ kg/mole ($\Delta M \sim 0.225$ kg/mole).

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